A first order assessment of the potential radiological impact of foodstuffs grown in a catchment area influenced by mining and mineral processing industries

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1. INTRODUCTION

Natural radioactivity is associated with the vast mineral resources in South Africa in such concentrations that the radionuclides from the natural uranium and thorium decay series are found to pose concern for public exposure to communities living around these areas. Consumption of water and food is usually the most important route by which natural radionuclides can enter the human body and assessment of natural radionuclide levels in different foods and diets is therefore important to estimate the intake of these radionuclides by man. In order to evaluate the yearly dose due to an individual source at a screening level of 25 μSv/a, one is faced with a required lower limit of determination (LLD) of 0.1 to 0.5 Bq/kg for certain foodstuffs

The aim of this study was to investigate the potential radiological impact of foodstuffs grown in a catchment area influenced by mining and mineral reprocessing industries in South Africa. Natural radionuclides were determined in a number of foodstuffs harvested from and/or grown in the area. The radionuclides were measured by non-destructive techniques such as Instrumental Neutron Activation Analyses (INAA) and low background gamma spectrometry. The estimated dose for the adult age group, resulting from consumption of these foodstuffs, was evaluated. The assessment of natural radionuclides in foods allowed us to evaluate the items that present the highest risk to the population, and compare this to the limits established by the National Nuclear Regulator (NNR).

2. MATERIALS AND METHOD

A variety of food samples from the six categories as defined by the World Health Organization i.e. cereals and grains, meat, seafood/fish, vegetables, roots and tubers, and fruit, were analyzed. The samples were collected in the vicinity of areas known to be influenced by mining and mineral processing industries. Samples were prepared as would apply for human consumption.

For determination of the U and Th element concentrations, sub-samples of approximately 200 mg each were weighed out in specially designed irradiation capsules and irradiated in the SAFARI-1 research reactor together with elemental standards. The uranium concentration (μg/g) was determined by delayed neutron counting. For determination of the thorium concentration (μg/g), the irradiated samples were cooled for 7 days and the 312 keV ^{233}Pa gamma-ray measured with a 25% relative efficiency well-type HPGe gamma detector.

A sub-sample of approximately 50 grams was analysed on a low-background high energy HPGe detector system with 20 % relative efficiency. The spectral data was analysed for the radionuclides ^{226}Ra, ^{228}Ra, ^{228}Th and ^{40}K. Samples were sealed and stored for 3 weeks before counting to allow for radium and its short-lived progeny to reach equilibrium.

For ^{210}Pb analyses, samples were adjusted to a specific counting geometry for analysis on a 10% relative efficiency coaxial, p-type HPGe gamma detector system with an ultra-thin beryllium window for low energy counting. Absorption corrections were made for each sample by counting a collimated absorption source. ^{210}Po was determined radiochemically after microwave digestion of 1 g of sample.

Since recently a low-background coaxial, p-type broad energy germanium detector with 50% relative efficiency, and much better sensitivity than the low and high energy counting systems, has been used for the analysis of food samples. Ashed samples of approximately 50 - 70 g were counted for 2 hours; dried samples were counted for 5-18 hours depending on the mass of sample available. The spectral data was analysed for ^{238}U, ^{226}Ra, ^{210}Pb, ^{235}U, ^{232}Th, ^{228}Ra and ^{228}Th.

3. RESULTS AND DISCUSSION

3.1 Activity concentrations in food samples

The analytical results for the natural radionuclides in the various dietary samples are presented in Table 1. As can be seen from the results a relatively large variability in radionuclide concentration was found for all types of food and even within the same type of food.

Many of the nuclides in all types of food samples had values below the LLD of the method employed. The fish samples had measurable concentrations for nuclides of the U series only, with ^{226}Ra slightly higher than ^{238}U and ^{210}Po. The activity concentrations of almost all the nuclides in meat were lower than the LLD. The concentration of ^{210}Po in the liver and kidney sample was significantly higher than the other nuclides. This can be expected since internal organs are known to accumulate ^{210}Po. A high concentration of ^{228}Th can probably be explained by the ingrowth of this radionuclide following the decay of ^{228}Ra, which is probably taken up in addition to the direct intake of ^{228}Th, rather than radioactivity from ^{232}Th.

In the vegetable crops a relatively large spread of concentrations was found, but it was lower than 0.5 Bq/kg for most of the vegetables. No significant difference was observed between the concentrations in leafy and root vegetables. The higher concentrations in spinach compared to the other leafy vegetables (cabbage and lettuce) suggests dominance of leaf deposition of nuclides from the atmosphere. Spinach requires minimal preparation prior to cooking, whereas the outer leaves of cabbage are usually removed, resulting in lower concentrations. The concentration in vegetables may vary seasonally, and the values in different years could also be variable. Meaningful estimates of dose would require continual sampling of crops throughout the year.

Enhanced concentrations of ^{226}Ra and ^{228}Ra were observed in the lemon sample. Radium has a high bio-availability and can be concentrated in vegetables. Radium is probably translocated to areas of maximum plant growth such as root tips, terminal leaves and fruit, and then becomes permanently fixed in these parts.

The very high concentrations found in oats and rye samples are a cause of

concern. The high results may be partly due to adhesion of soil particles to the leaves.

3.3 Committed effective dose

Intake via ingestion depends on the dietary habits of the affected groups and/or individuals (which is also age related), and the concentration of the radionuclides concerned. The annual intake was estimated by multiplying the radionuclide concentration in the food by the amount of food consumed per year. Doses were evaluated using the dose conversion factor for ingestion exposure for (Sv/Bq), recommended by the ICRP. The overall estimated yearly dose based on the additional information as explained underneath, is reported in Table 1. In the absence of data specific to South African conditions, default consumption values for foodstuffs as applied by the South African National Nuclear Regulator (NNR) were used.

A conservative approach has been followed to obtain the yearly dose if the individual food source(s) would be used as the only food source available to the adult population. Equilibrium with the respective mother/daughter nuclides were assumed for those radionuclides not analysed for (^{210}Pb = ^{210}Po; ^{238}U = ^{234}U = ^{230}Th). For the short-lived nuclides such assumptions are acceptable, but for nuclides such as ^{227}Ac and ^{231}Pa these assumptions are not entirely accurate. Assumption of equilibrium between the two uranium isotopes can probably be expected for the samples obtained from the catchment area where uranium is the dominant contributor in the effluents from the mining and mineral processing operation. Assumed equilibrium between ^{210}Po and ^{210}Pb will result in an underestimation of the dose for those foodstuffs with Po:Pb ratios much lower than 1. Equilibrium can only be assumed if the chosen vegetable is going to be stored for one or more years (e.g. canned foods). In essence the activity of ^{210}Po, ^{230}Th, ^{231}Pa and ^{227}Th should also be determined, as there is no reason to assume that they could not be available in excess to their mother nuclides.

By ignoring the contribution of radionuclides measured lower than the LLD, especially where the LLD is quite high, the dose may be underestimated. Therefore values lower than the LLD is taken as LLD/2 in the dose calculations. However, if the radionuclides are not measured with the necessary sensitivity, the dose will most likely be overestimated. This makes it clear that this generally accepted practice in environmental monitoring should be applied with great care not to produce meaningless results and/or overestimate the results. One should rather develop/use adequately sensitive (radio) analytical techniques to estimate the dose to members of the public more accurately. Table 2 shows the theoretically calculated LLDs of the instrumental methods required to measure the different radionuclides with the necessary sensitivity to evaluate the dose at a level of 25 µSv/a. The low LLD required for root vegetables is as a result of the high consumption rate. From Table 2 it is clear that for most of the nuclides the necessary sensitivity can be obtained by analysing ashed samples on the broad energy detector. Due to the high dose conversion factors of ^{210}Po, ^{230}Th, ^{231}Pa and ^{227}Th, these nuclides have to be analysed radiochemically, since the sensitivity of the instrumental techniques is inadequate to obtain the required LLD.

For the majority of the analysed foodstuffs the estimated dose was less than 250 µSv/a. The higher dose obtained for some samples indicated in bold in Table 1, (e.g. maize, spinach, beetroot) is probably overestimated because of high LLDs for ^{238}U (= ^{230}Th). The high dose for meat was due to the high LLD for ^{210}Pb obtained by low-energy gamma spectrometry. Assuming equilibrium with ^{210}Po, the dose decreases to 0.144 µSv/a. For liver the dose was dominated by ^{210}Pb and

^{210}Po, with ^{210}Po contributing almost 80% of the overall dose. It must be noted that an unrealistic high consumption rate of 100 kg per annum used in the dose assessment, contributes to the high calculated dose. High concentrations of ^{228}Ra and ^{226}Ra contribute to the relatively high observed dose for the lemon samples.

For most of the foodstuffs ^{210}Po was the most important contributor to the overall dose, whereas the contribution from ^{238}U and ^{232}Th was small (less than 10 %). A graphical distribution of the dose per nuclide for the adult age group, as found for the majority of the samples, is shown in Figure 1. The contribution of ^{210}Po to the overall dose was about 20 - 40%; other main contributors were ^{228}Ra , ^{226}Ra and ^{210}Pb. For some samples ^{230}Th also contributed significantly, not necessarily because of actual high measured activity concentrations but mainly because of the high dose conversion factors of ^{230}Th, and assumed equilibrium with ^{238}U which was measured with poor sensitivity (e.g. maize, spinach 2 and beetroot 2 samples). As shown in the graphical distribution for the beetroot sample (Figure 2) this "overestimated" contribution was as high as 40%.

The main items of concern therefore seem to be the oats 2, rye and lucern samples. The samples indicated as "*fodder*" in Table 1, represent samples of materials used as animal feed. The calculated doses should therefore not be done for human consumption, but via the animal pathway. The resulting doses via meat and milk were higher than 1 mSv/a, and the possibility of the samples being contaminated can therefore not be ignored. As mentioned earlier, it is important to note that one cannot make decisions based on the analysis of one sample of a specific foodstuff. These particular samples were taken in the dry season and it is most likely that the results may differ substantially if sampled in the wet season when irrigation requirements are reduced. Food processing can also greatly reduce the radioactive content of final edible products; neglecting the losses during food processing can lead to an overestimation of the calculated dose.

The annual effective dose (excluding the individual samples with high dose indicated in bold in Table 1) varies between 25 and 170 µSv/a. Compared to the annual effective dose of no more than 200 µSv/a from natural radionuclides in food as assessed by UNSCEAR this does not imply an unacceptable level of exposure.

The NNR Guide LG-1032 refers to an annual screening assessment criterion of 25 µSv for a specific pathway, taking into consideration also the contribution from other pathways, so that an overall criterion of 250 µSv is followed. From the evaluation provided in Table 1, it can be seen that for most of the samples, the calculated doses for adults exceed the screening criteria of 25 uSv, but are less than the overall criteria of 250 µSv due to a specific source. The individual samples which exceed the maximum exposure level of members of the public set at 1 mSv/a is of obvious concern and further investigation is mandatory.

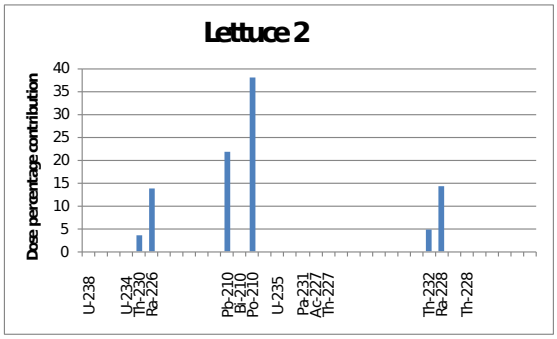

Figure 1. Dose contribution from individual nuclides for the adult age group for Lettuce (representing the majority of the vegetable samples)

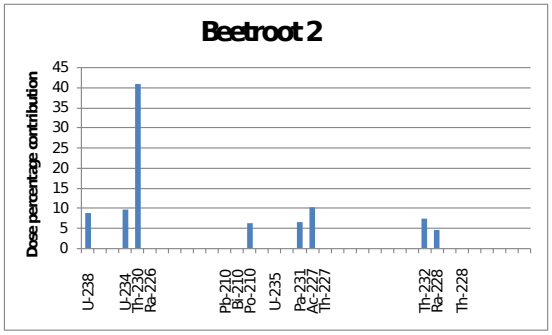

Figure 2. Dose contribution from individual nuclides for the adult age group for Beetroot 2 (illustrating overestimation due to high LLD for ^{238}U (= ^{230}Th))

4. CONCLUSIONS

It was shown that measurement of ashed samples by broad energy gamma spectrometry greatly improves the LLD of the direct instrumental techniques, thereby providing suitable data for evaluation of the yearly dose at the screening level of 25 µSv/a. However, nuclides such as ^{230}Th, ^{210}Po and ^{231}Pa has to be analysed by radiochemical separation through acid destruction of dried foodstuffs followed by individual element separations to provide suitable data. Overestimation of dose due to poor measurement detection limits, was clearly indicated for some of the samples.

Exposure from intakes of foods with these activity concentrations would lead to individual annual dose of not more than 0.25 mSv, which does not imply an unacceptable level of exposure. The results from this study therefore do not provide any indication of a significant impact of the mining activities on food grown in the area. Apparent high doses from individual samples with high activities, were mostly overestimated because of high LLDs. Oats, rye and lucern samples were considered as a potential risk which could result in public exposure up to a few mSv per year, if used as animal feed and considering the fodder-animal-human pathway. The possibility of contamination in the area can therefore not be ignored.

This study is considered as a first order assessment of the potential radiological impact on the public in the study area, since extrapolation based on single samples is unlikely to be reliable. For a rigorous estimate of actual yearly dose the following should be considered: (a) More frequent sampling monitored over a longer period, (b) Application of real, more accurate, consumption/intake values, and (c) Verification of equilibrium assumptions or preferably, measurement of the specific nuclides of concern e.g. ^{230}Th.

Table 1. Activity concentration of radionuclides in Bq/kg wet weight and Committed Effective dose for the adult age groups (mSv/a)
**Analysis by broad energy gamma spectrometry on ashed samples*

Category	U-238	Ra-226	Pb-210	Po-210	U-235	Th-232	Ra-228	Th-228	Dose
Fish 1 *	0.66 ± 0.13	1.4 ± 0.1	0.24 ± 0.09	0.24 ± 0.09	0.031 ± 0.006	< 0.044	< 0.38	0.025 ± 0.004	0.031
Meat	< 3.3	< 0.58	**< 17**	0.13 ± 0.047	< 0.15	< 0.45	0.82 ± 0.39	< 0.64	**0.752**
Liver &	0.20 ± 0.11	< 2.7	1.2 ± 0.48	3.87 ± 0.49	0.0091 ± 0.0051	< 0.30	< 0.9	0.341 ± 0.040	**0.603**
Kidney									
Grains &									
Cereals	**< 35**	< 2.8	-	0.28 ± 0.141	< 1.6	**< 6**	< 5	**< 13**	**1.67**
Maize									
Vegetables									
Spinach 1	2.29 ± 0.24	< 0.27	0.435 ± 0.083	0.44 ± 0.083	0.106 ± 0.011	0.075 ± 0.073	0.305 ± 0.032	0.126 ± 0.009	0.111
Spinach 2	**< 30**	1.0 ± 0.3	0.762 ± 0.155	0.65 ± 0.103	< 1.4	3.93 ± 0.64	< 1.3	0.82 ± 0.25	**0.488**
Spinach 3*	0.289±0.027	< 0.84	1.41 ± 0.023	-	0.0133 ± 0.0013	0.40 ± 0.15	< 0.35	0.153 ± 0.014	0.170
Cabbage 2*	0.242±0.026	< 0.60	< 0.35	-	0.0111 ± 0.0012	0.081 ± 0.038	< 0.24	< 0.062	0.034
Lettuce 1*	0.0791±0.0178	< 0.36	< 0.24	-	0.00364±0.00082	0.14 ± 0.039	< 0.38	0.0279 ±0.0049	0.025

Roots & Tubers									
Turnips 1	0.320 ± 0.123	< 0.16	< 0.14	-	0.0147 ± 0.0057	0.017±0.0025	< 0.088	0.021 ± 0.005	0.051
Turnips 2*	0.0848±0.0179	< 0.66	< 0.42	-	0.00390±0.00083	< 0.23	< 0.23	< 0.069	0.108
Beetroot 1		< 0.21	0.255 ± 0.065	0.26 ± 0.065		0.075±0.0039	0.174 ± 0.021	0.122 ± 0.006	0.162
Beetroot 2	0.714 ± 0.191	0.46 ± 0.19			0.0329 ± 0.0088		< 0.72	< 0.44	
Spring Onions		0.78 ± 0.02	0.147 ± 0.053	0.28 ± 0.053		3.0 ± 0.9	0.27 ± 0.07	0.37 ± 0.02	**0.917**
Carrots*	**< 21**	< 0.59	0.61 ± 0.13	0.60 ± 0.13	< 0.98	0.073 ± 0.003	< 0.23	0.215 ± 0.014	0.097
Sweet Potatoes*	00.34 ± 0.12	< 0.38	< 0.37	-	0.0170 ± 0.0060	0.195 ± 0.053	< 0.16	0.052 ± 0.0055	0.104
	0.0588±0.0158		< 0.27	0.43 ± 0.09	0.00271±0.00073	0.028 ± 0.003			0.127
	0.029 ± 0.0057				0.00133±0.00026				
Fruit									
Lemon	< 0.61	**2.94 ± 0.63**	-	0.14 ± 0.038	< 0.028	0.13 ± 0.05	**3.83 ± 0.83**	< 2.8	**0.305**
Fodder									
Oats 1	< 0.27	0.39 ± 0.28	8	1.39 ± 0.339	<0.013	< 0.034	< 0.012	0.012 ± 0.029	0.421
Oats 2	**316 ± 27**	1.4 ± 0.3			**14.6 ± 1.3**	**6.65 ± 0.53**	< 3.2		**21.5**
Rye	< 8.5	**14.5 ± 0.9**	**5.05 ± 0.65**	**5.64 ± 0.360**	< 0.39		< 16	**2.28 ± 0.62**	**4.75**
Barley Leaves	< 2.6	**11 ± 5**	**9.82 ± 1.30**	**9.55 ± 0.927**	< 0.12	0.39 ± 0.09	< 3.5	**27.3 ± 9**	0.183
Lucern	1.90 ± 1.21	< 1.9	-	0.38 ± 0.110	0.087 ± 0.010	0.387 ± 0.053	**3.99 ± 0.68**	6.9 ± 2.3	
		3.80 ± 0.44		0.50 ± 0.083		0.217 ± 0.033		< 1.9	**0.855**

Table 2 : LLD (Bq/kg) required for a dose of 25 µSv/a based on consumption rates and dose conversion factors as indicated

*Refers to LLD of radiochemical method for ^{210}Po; Values in brackets are average experimental LLDs for the vegetable group as obtained from Table 1

Nuclide	DCF adult (Sv/Bq)	Root veg	Leafy veg	Fruit	Cereal	Fish	Chicken	Meat	Theoretical LLD: wet weight (average experimental LLDs in brackets)		
Consumption (kg/a)		170	55	75	150	25	75	100	NAA/high gamma	Broad gamma dried	Broad gamma Ashed
U-238	4.50E-08	3.27	10.1	7.41	3.70	22.2	7.41	5.56	6 (> 1)	1.2 (~0.1)	0.034 (~ 0.05)
U-234	4.90E-08	3.00	9.28	6.80	3.40	20.4	6.80	5.10	6	1.2	0.034
Th-230	2.10E-07	0.70	2.16	1.59	0.79	4.76	1.59	1.19			
Ra-226	2.80E-07	0.53	1.62	1.19	0.60	3.57	1.19	0.89	2 (~ 0.8)	0.34 (~ 0.6)	0.03 (~0.3)
Pb-210	6.90E-07	0.21	0.66	0.48	0.24	1.45	0.48	0.36		1.7 (~0.6)	0.33 (~ 0.2)
Po-210	1.20E-06	0.12	0.38	0.28	0.14	0.83	0.28	0.21	*0.05 (~0.2)		
U-235	4.70E-08	3.13	9.67	7.09	3.55	21.3	7.09	5.32	0.2 (~ 0.5)	0.18 (~ 0.02)	0.0015 (~0.002)
Pa-231	7.10E-07	0.21	0.64	0.47	0.23	1.41	0.47	0.35			
Ac-227	1.10E-06	0.13	0.41	0.30	0.15	0.91	0.30	0.23			
Th-232	2.30E-07	0.64	1.98	1.45	0.72	4.35	1.45	1.09	3 (> 1)	0.7	0.054 (~ 0.2)
Ra-228	6.90E-07	0.21	0.66	0.48	0.24	1.45	0.48	0.36	2 (> 1)	0.7	0.11 (~ 0.4)

Th-228	7.20E-08	2.04	6.31	4.63	2.31	13.89	4.63	3.47	2 (> 1)	0.2 (~0.07)	0.026 (~ 0.03)